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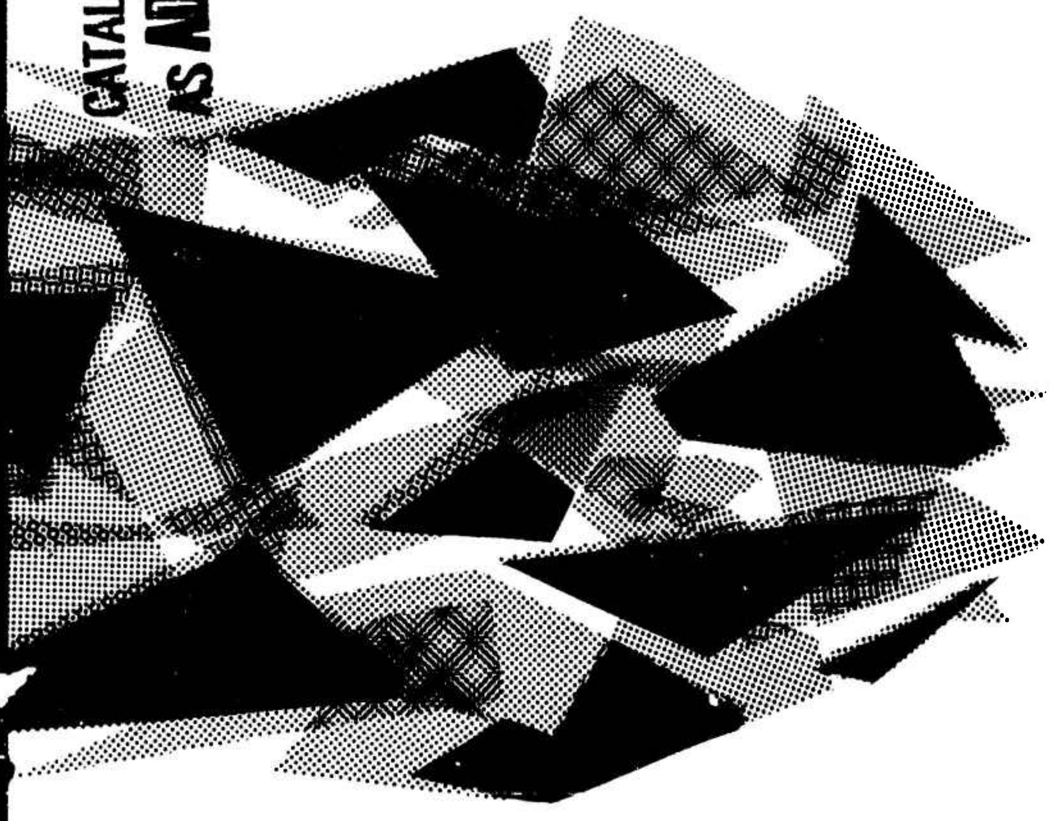
REDSTONE ARSENAL RESEARCH DIVISION
HUNTSVILLE, ALABAMA

SPECIAL REPORT NO. S-67

FAILURE DIAMETER STUDIES OF COMPOSITE PROPELLANTS
I. Diameter-Dependence of Detonation Velocities of Explosives-Loaded Formulations (U)



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FAILURE DIAMETER STUDIES OF COMPOSITE PROPELLANTS

I. Diameter-Dependence of Detonation Velocities of Explosives-Loaded Formulations

by

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FAILURE DIAMETER STUDIES OF COMPOSITE PROPELLANTS

I. Diameter-Dependence of Detonation Velocities of Explosives-Loaded Formulations

ABSTRACT

Work with active-binder propellants is described which has shown that failure diameters may be predicted from measurements of D_f and detonation velocity of RDX-adulterated formulations. Application to fuel-binder propellants has been attempted but the exact relations successful with the earlier types do not hold. In the latter case, experimental results indicate that in RDX-loaded fuel-binder composite propellants only the RDX is responsible for detonation and that the base propellant is nondetonable. Marked differences among failure diameters determined by direct-match and so-called inverse techniques have been found which have serious implications as regards hazard evaluation of very large solid propellant motors.

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FAILURE DIAMETER STUDIES OF COMPOSITE PROPELLANTS

I. Diameter-Dependence of Detonation Velocities of Explosives Loaded Formulations

Few studies of the detonability of fuel-binder solid propellants have been conducted, because these substances do not detonate in charge sizes which can be handled conveniently. Several methods are available by which failure diameter can be reduced: introduction of porosity, increased confinement, and addition of high explosive. This Division, having developed a means of predicting failure diameters of reactive-binder propellants from failure diameters and detonation velocities of RDX-adulterated formulations, has undertaken, for the Air Force, to apply similar techniques to non-reactive binder propellants.

BACKGROUND

Derivation of Extrapolation Method[1*]

The method of extrapolation comes from a consideration of the kinetics of the system. If the rate-limiting reaction in the detonation process is a single first-order step, the reaction time is given by [2]

$$t = \frac{c_v RT^2}{\nu E_a Q} \exp\left(\frac{E_a}{RT}\right) \tag{1}$$

The reaction time can be related to the detonation velocity and charge diameters through the "curved front" theory of Eyring et al. [3]

$$\frac{U^o - U}{U^o} = \frac{\xi_1}{D} \tag{2}$$

and their assumption that

$$\xi_1 = 0.75tU \tag{3}$$

*Superscripts indicate references listed at end of report.

Combination of Eqs. 1 through 3 gives

$$\frac{D}{0.75\ U}\left(\frac{U^{o} - U}{U^{o}}\right) = \frac{c_{v}RT^{2}}{\nu E_{a}Q}\ exp\left(\frac{E_{a}}{RT}\right) \tag{4}$$

or, for the particular case of the failure diameter,

$$\frac{D_{f}}{0.75\ U_{f}}\left(\frac{U^{o} - U_{f}}{U^{o}}\right) = \frac{c_{v}RT^{2}}{\nu E_{a}Q}\ exp\left(\frac{E_{a}}{RT}\right) \tag{5}$$

In non-reactive binder propellants the reaction times are so long that failure diameters become very large. Addition of RDX, which rapidly releases a large amount of energy, shortens the reaction time by raising Q in Eq. 5.

If addition of a high explosive does not alter the rate-determining step in the detonation reaction of the propellant, the temperature T which must be attained for detonation to occur remains the same, as do E_{a} and ν. If, at the same time, the physical properties of the propellant do not change signifícnatly, c_{v} is also constant. Eq. 5 can then be reduced to

$$D_{f}\left(\frac{U^{o} - U_{f}}{U^{o}}\right) = \frac{AU_{f}}{Q} \tag{6}$$

where

$$A = \frac{0.75\ c_{v}RT^{2}}{\nu E_{a}}\ exp\left(\frac{E_{a}}{RT}\right)$$

A is a constant for any individual base propellant.

In this equation Q is the combined heat of detonation for the base propellant and the added explosive

$$Q = n_{1}Q_{1} + n_{2}Q_{2} \tag{7}$$

where

n_1 = weight fraction of explosive

n_2 = weight fraction of base propellant

Q_1 = heat of detonation of explosive per gm

Q_2 = heat of detonation of base propellant per gm

The velocities U^o and U_f in Eq. 6 can be determined from measurements of velocity as a function of diameter. The data are plotted with U as a function of 1/D and an extrapolation is made to infinite diameter and failure diameter. Measurements of velocity as a function of diameter at two RDX concentrations give the values of A and Q_2 for a base propellant formulation. Q_1 is assumed to be the heat of explosion of RDX.

When A and Q_2 have been determined, the only quantity still unknown in Eq. 6 is $(U^o - U_f)/U^o$. The velocities can be calculated from hydrodynamic theory but, since the expression involves a small difference between two large numbers, calculated velocities are not accurate enough. A relationship between $(U^o - U_f)/U^o$ ratio and some property of the base propellant is needed. Such a relationship has been found in plastisol-nitrocellulose composite propellants.

Determination of $(U^o - U_f)/U^o$ for Plastisol Propellants

A series of velocity vs. diameter determinations has been made for several ammonium perchlorate-containing plastisol-nitrocellulose propellants[4].(Table I). One of these compositions contained aluminum and no RDX, one had neither aluminum nor RDX and the other three contained RDX and no aluminum. Using these data the following empirical relation was evolved:

$$-\log\left(\frac{U^o - U_f}{U^o}\right) = 0.0415\ D_f + 0.990 \quad [D_f \text{ in cm}] \qquad (8)$$

Table I

Detonation Parameters for Non-Aluminized Plastisol Propellants Used to Determine Constants in Eq. 7

Propellant	% RDX[a]	D_f, in.	U^o, mm/μsec	$(U^o - U_f)/U^o$
RH-P-112[b]	0	1.05 - 1.15	6.407	0.081
RH-P-181[c]	0	1.82 - 1.92	6.598	0.066
RH-P-184	5.9	1.25 - 1.50	6.702	0.069
RH-P-223	15.0	1.00 - 1.05	7.060	0.085
RH-P-205	35.3	0.36 - 0.50	7.309	0.090

[a] substituted for ammonium perchlorate
[b] contains 15% Al
[c] RH-P-112 minus aluminum; base for succeeding formulations

The three RDX-containing compositions (RH-P-184, RH-P-223 and RH-P-205) were used to calculate the failure diameter of the base propellant, RH-P-181. The value of D_f was calculated to be 1.96 in.; the measured value was 1.82 in. $< D_f <$ 1.92 in.

Another series of failure diameter measurements had been made for an aluminized, ammonium perchlorate-containing, plastisol-nitrocellulose composition with varying amounts of RDX[5] (Table II). Since no velocity data had been determined, velocities were assumed from those of the other series. The failure diameter of the base propellant was calculated to be 1.26 in.; the measured value was 1.05 in. $< D_f <$ 1.38 in.

Table II

Failure Diameters for Aluminized Plastisol Formulations with Varying RDX Content (Ref. 5)

Formulation	% RDX[a]	D_f, in.
RH-P-116	0	1.00 - 1.25
RH-P-144	2	0.82 - 1.05
RH-P-145	5	0.65 - 0.82
RH-P-135	10	0.36 - 0.62
RH-P-146	20	<0.36

[a]substituted for ammonium perchlorate

PROGRAM

Despite the success of the foregoing method, it possesses little practical use with reactive-binder propellants since their detonation characteristics can easily be examined on unadulterated forms. The real value of this method would be in its successful application to fuel-binder formulations.

The program to test the applicability to non-reactive-binder composite propellants was laid out in two phases. If Eq. 8 applies to all binder systems, velocity measurements are needed for only two RDX-containing compositions (Phase I). However, if Eq. 8 is applicable only to a particular type of binder, then several RDX concentrations must be studied to redetermine the form of the relationship between D_f and $(U^o - U_f)/U^o$, if such exists (Phase II).

APPLICATION TO PBAA PROPELLANTS

Experimental

Two polybutadiene-acrylic acid propellant formulations containing RDX were selected (see Appendix A) for velocity determinations. The base formulation, RH-B-15, was composed of 62.5 weight per cent of a bimodal blend, 70% 180μ and 30% 15μ wt.-median-diameter ammonium perchlorate; 12.5 weight per cent 3 to 5μ aluminum, and a PBAA/ERL2774 = 89.5/10.5 binder. To this base formulation Type B Class E RDX (97%<44μ; 17μ wt.-median-diam. [Micromerograph]*) was added to make formulations RH-B-17 and RH-B-25, which contained 25 and 18 weight per cent RDX, respectively; failure diameters alone were determined for two other formulations containing 16% and 20% RDX, RH-B-23 and RH-B-18, respectively (Table III). The RDX-containing formulations were cast into wax-coated cardboard tubes and cured for four days at 140°F. The velocity charges so obtained were 30 to 40 inches long and 1.25 to 6 inches in diameter. Control charges were cast from each batch to assure that the failure diameter of the propellant being examined was constant (Table IV). This was necessary since not all the charges could be cast from the same batch. The batch size was about 45 pounds; in the case of the larger charges, this limitation determined the length of the velocity rounds.

Table III

Propellant Formulations

Ingredient, Wt. %	RH-B-15	RH-B-17	RH-B-18	RH-B-23	RH-B-25
APC	62.5	46.9	50.0	52.5	51.3
Al	12.5	9.4	10.0	10.5	10.2
PBAA/ERL 2774[a]	25.0	18.7	20.0	21.0	20.5
RDX	----	25.0	20.0	16.0	18.0

[a]Epoxy resin, Bakelite Div., Union Carbide Corp.

*See Appendix A.

Table IV

Failure Diameters of RDX-Loaded PBAA Formulations

Designation	Batch	% RDX	D_f, in.
RH-B-17	1002	25	1.00 - 1.25
	1003		1.00 - 1.25
RH-B-25	1000	18	2.25 - 2.50
	1001		2.25 - 2.50
	1002		2.25 - 2.50
	1003		2.25 - 2.50
	1006		> 2.5
	1007		2.25 - 2.50

The detonation velocities of the propellants were determined by measuring the time intervals between ion probes[6] inserted at five equally spaced positions along the axis of the charge. The detonation of each round was initiated at one end by means of a Composition C-4 booster having a bulk density of 1.59 gm./cc. and a diameter equal to that of the charge; the 6-inch-diameter velocity charge was initiated with a 4-inch-diameter booster. The boosters had length-to-diameter ratios of 3; they were cylindrical in shape through 2.5-inch diameters and conical at the larger sizes. Initiation of the booster was by an Engineers Special electric blasting cap, J2.

The ion probes were fabricated at this Division from 19-gage stainless steel tubing normally used in the manufacture of hypodermic needles. Within these were inserted 24-gage enameled magnet wire, secured in place by Duco cement. The steel sheath was partially cut away from the sensing end to expose the inner conductor; the tip of the assembly was beveled to permit easy insertion into the propellant charge (Fig. 1). After initial development, these probes proved to have excellent reliability and loss of information at a station was rare.

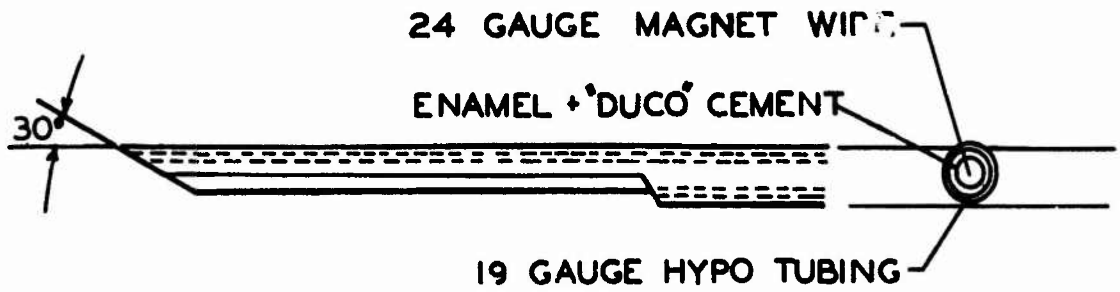

FIG. 1 ION-PROBE ASSEMBLY.

One ion probe, serving as a trigger, was placed between the booster and charge. Five additional probes were located at equally spaced stations down the charge, the first at 3 inches from the interface. Each downstream probe led to an 8-megacycle counter chronograph (Potter); circuitry was so arranged that the counter would be stopped upon the probe's becoming conductive (Fig. 2). The chronographs were started in common by the trigger and the incremental times were obtained by difference. This arrangement was made redundant with the use of three 10-megacycle interval timers (Beckman) capable of resolving 0.1 μsec. The ion probes to the two sets of counters shared a common power supply, but the systems were otherwise independent, each set of counters having its own trigger.

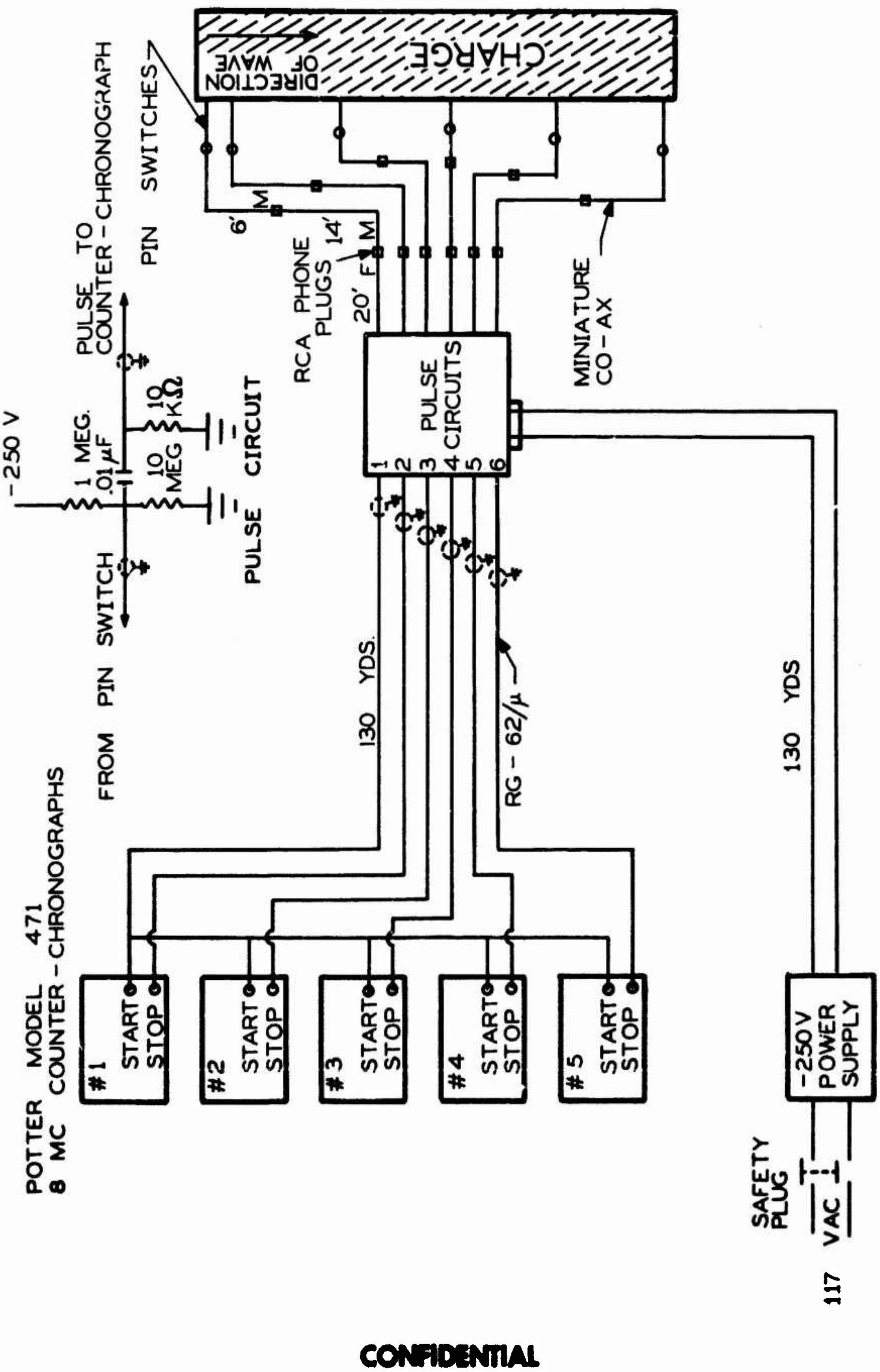

FIG. 2 VELOCITY-MEASUREMENT CIRCUITRY.

Detonation velocities were determined from the harmonic mean of the velocities over successive increments. The intermediate stations also served as indications of the stability of the detonation velocity. The layout of the ion probes in the charges of interest is shown in Figs. 3-5. The detailed distance—time data for all shots are given in Table A-IV of Appendix A.

The failure diameter D_f was established, within limits, for each batch of propellant cast by initiating propellant charges ($L/D \geqq 4$) with Composition C-4 boosters of matching diameters; the occurrence of detonation was determined by perforation of a mild steel witness plate. It was found that, with PBAA-type propellant, the witness plate did not always give an unambiguous result, i.e. the plate was not always cleanly punched. When this occurred the initiation of a duplicate round was photographed with a streak camera (Beckman & Whitley Model 194). The criterion of detonation was then a luminous front of constant velocity. The failure diameter of RH-B-17 (25% RDX) was found to be between 1.00 and 1.25 inches and that of RH-B-25 (18% RDX) between 2.25 and 2.50 in. (Table IV). Since, with the latter formulation, the witness plates were not always cleanly punched, the failure diameter was inferred to be closer to 2.50 in. than to 2.25 in.

Results of Measurements on PBAA Propellants

Detonation velocities were measured over nearly a 3-fold charge-diameter range for PBAA propellant containing 18% and 25% RDX (Table V). The velocity data plotted as a function of 1/D (Fig. 6) lay along straight lines described by

$$\frac{U}{5.738} = 1 - \frac{0.0448}{D} \qquad (9)$$

for 25% RDX and

$$\frac{U}{5.358} = 1 - \frac{0.117}{D} \qquad (10)$$

for 18% RDX.

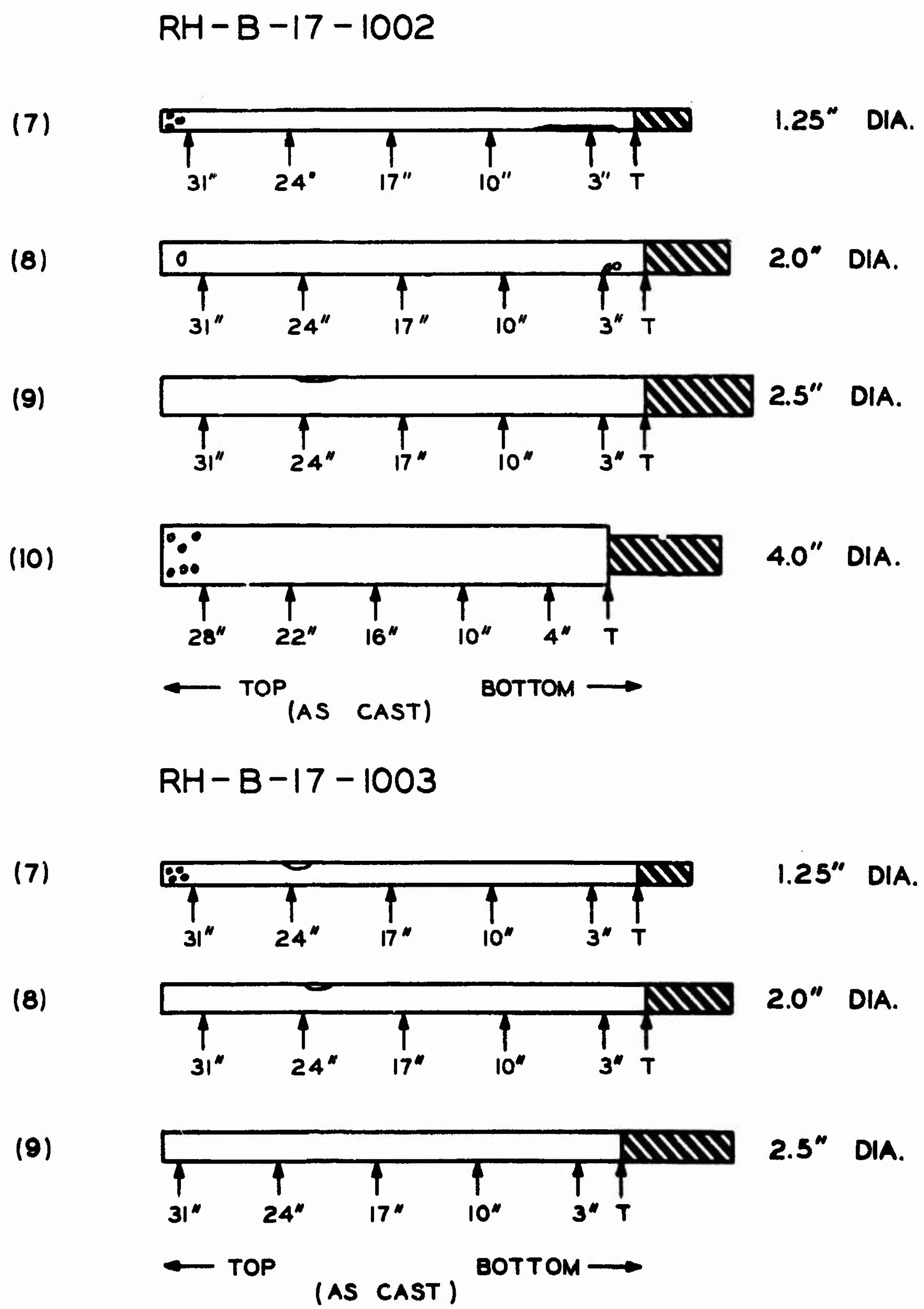

FIG. 3 LAYOUT OF CASTING IMPERFECTIONS AND ION-PROBE STATIONS IN RH-B-17 VELOCITY CHARGES.

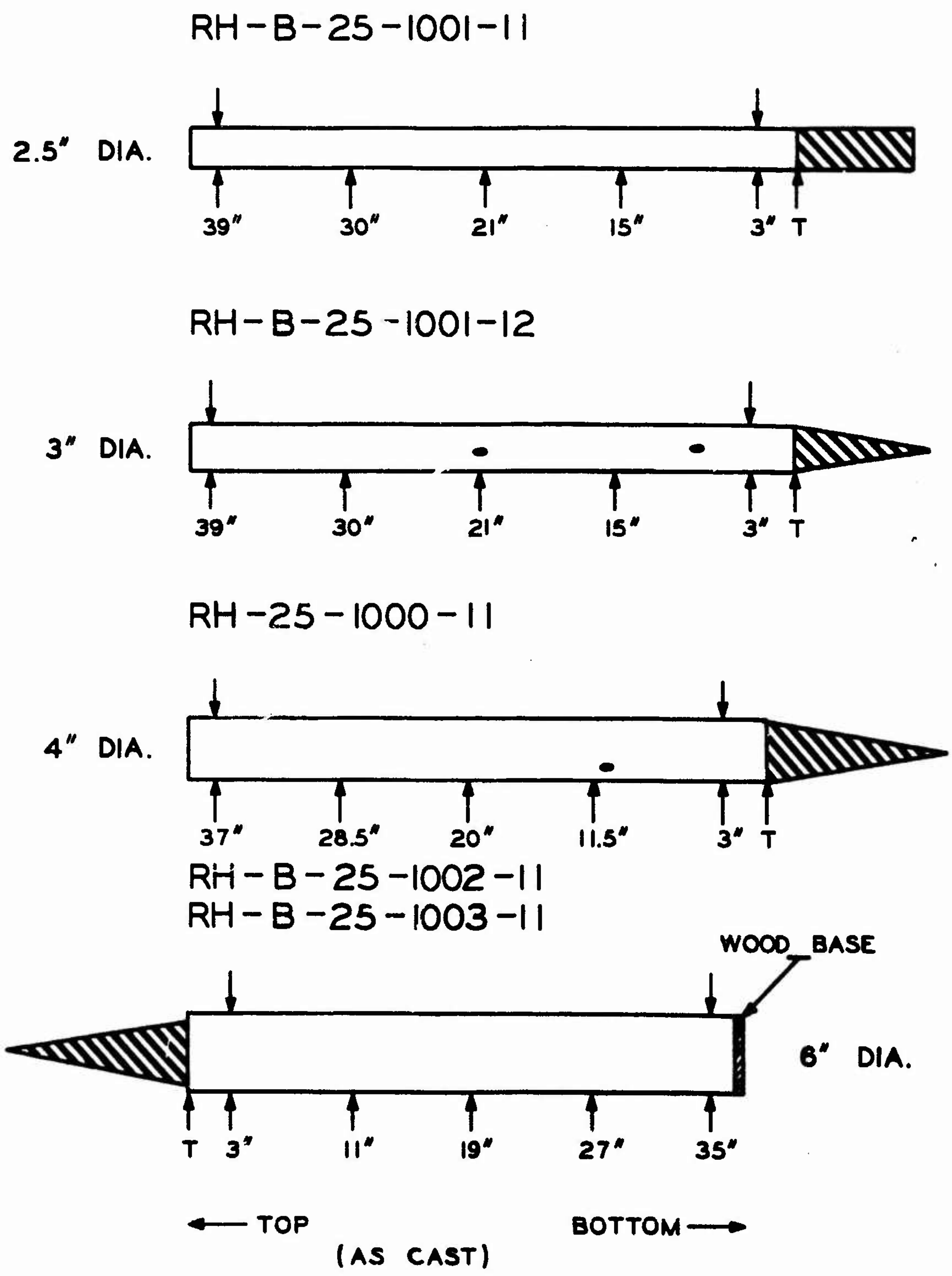

FIG. 4 LAYOUT OF CASTING IMPERFECTIONS AND ION-PROBE STATIONS IN RH-B-25 VELOCITY CHARGES.

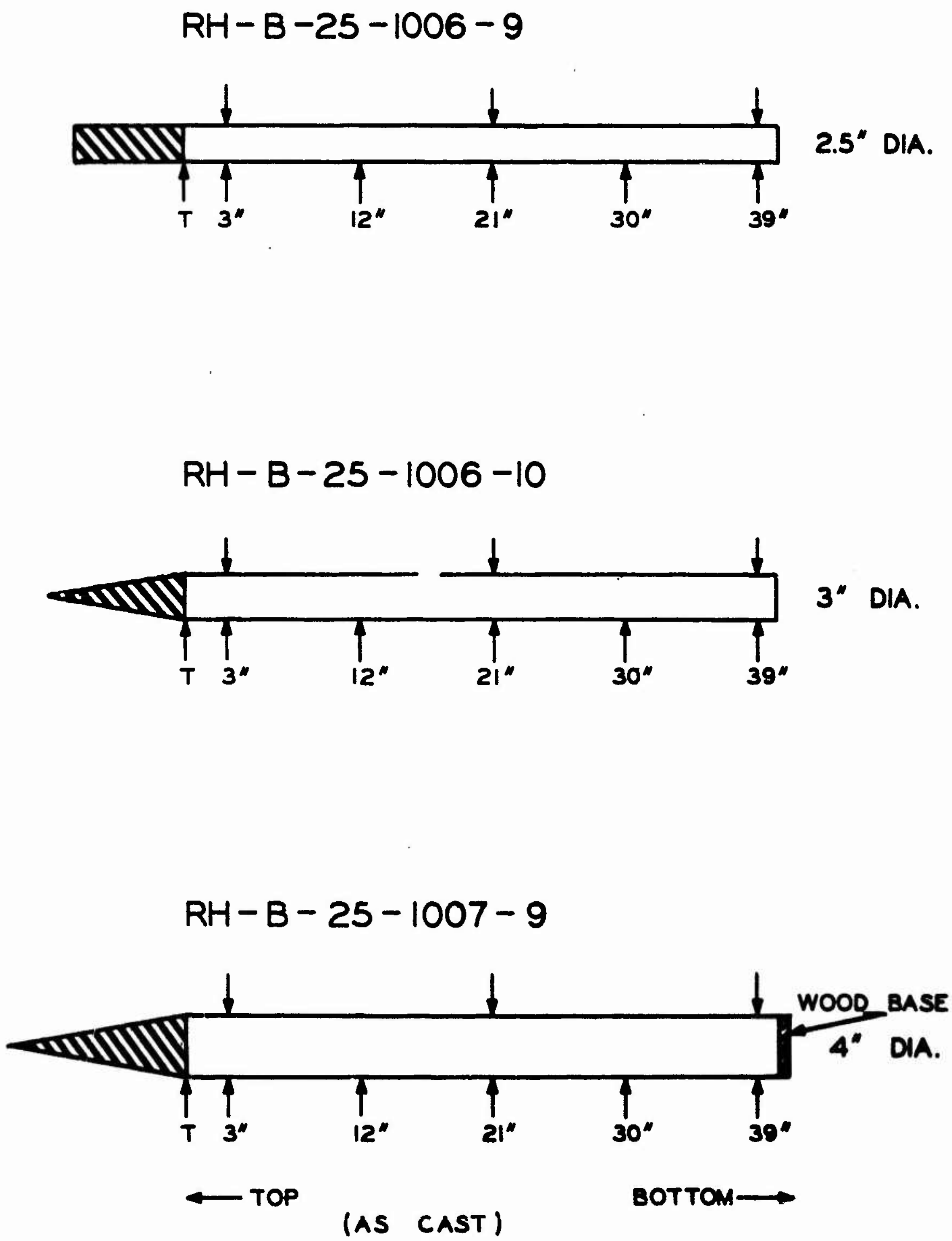

FIG. 5 LAYOUT OF CASTING IMPERFECTIONS AND ION-PROBE STATIONS IN RH-B-25 VELOCITY CHARGES.

Table V

Velocity vs. Diameter Data for PBAA Propellants Containing RDX

% RDX	D_f, in.	Charge Diam., in.	Velocity (mm/µsec)
25	1.00 - 1.15	1¼	5.519
25		1¼	5.531
25		2	5.594
25		2	5.644
25		2½	5.633
25		2½	5.622
25		4	5.666
18	2.25 - 2.5	2½	5.116
18		3	5.144
18		3	5.137
18		4	5.222
18		4	5.188
18		6	5.252

The detonation parameters were calculated from Eqs. 9 and 10 (Table VI); 2.5 in. was used for D_f for the 18%-RDX composition and 1.08 in. for the 25%. From these values and by assuming a relation of the same type as Eq. 8 the following equation was obtained

$$-\log\left(\frac{U^o - U_f}{U^o}\right) = 1.4178 - 0.0352\, D_f \quad [D_f \text{ in inches}] \qquad (11)$$

Solving for the constants in Eq. 6 gave Q_2 = -210 cal/gm and A = 1.325

$$D_f\left(\frac{U^o - U_f}{U^o}\right) = \frac{1.325\, U_f}{1280 n_1 - 210 n_2} \qquad (12)$$

This equation was used to draw plots of $(U^o - U_f)/U^o$ vs. D_f for several RDX concentrations.

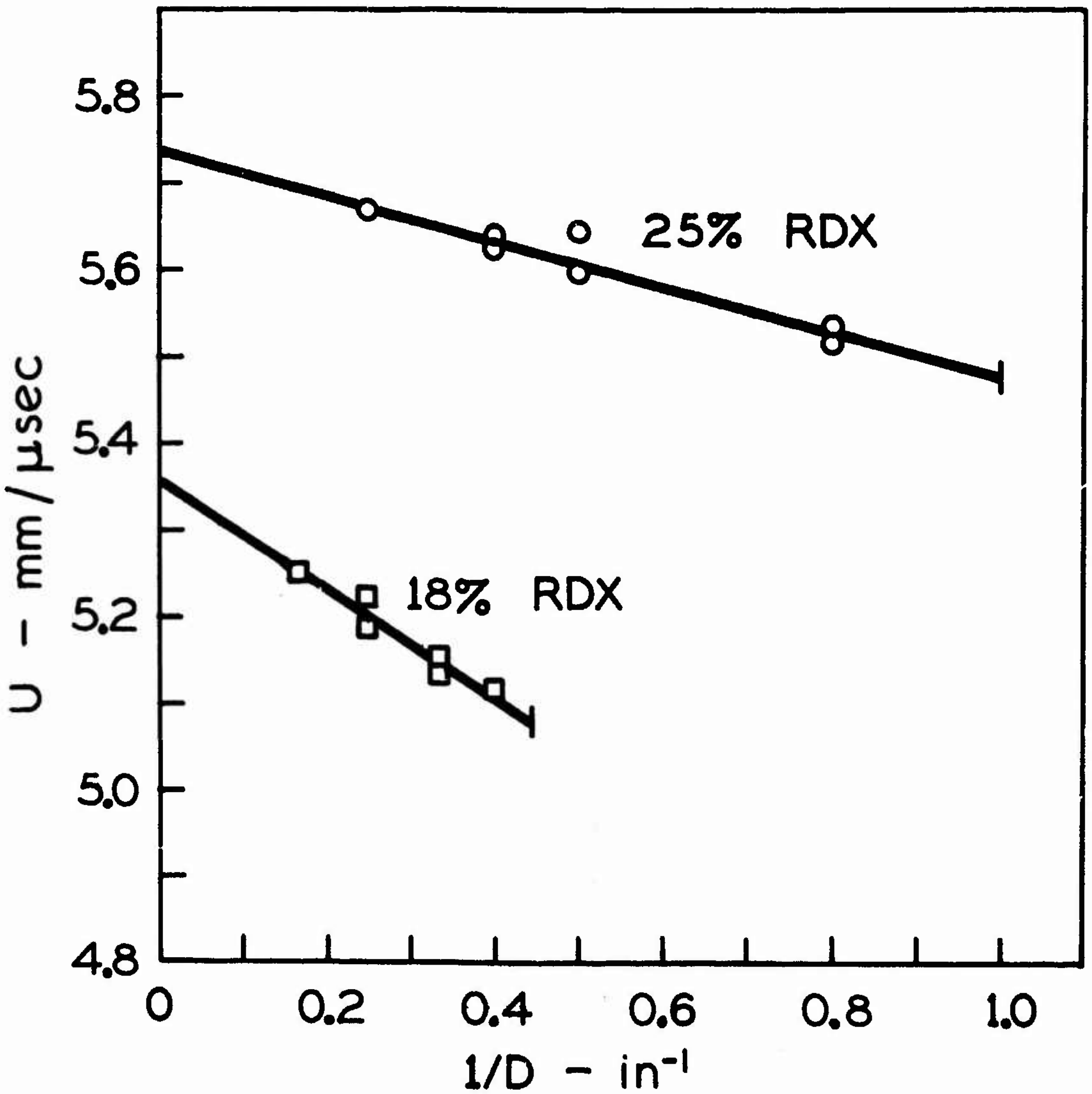

FIG. 6 DIAMETER-DEPENDENCE OF DETONATION VELOCITY OF PBAA COMPOSITE PROPELLANTS CONTAINING RDX.

Table VI

Detonation Parameters for RDX-Loaded PBAA Propellant

% RDX	D_f, in.	U^o (mm/µsec)	U_f (mm/µsec)	$(U^o - U_f)/U_f$
25	1.00 - 1.15	5.735	5.496	0.0417
18	2.25 - 2.5	5.358	5.107	0.0468

The failure diameter of a particular formulation is given by the intersection of its curve with that of Eq. 11. Fig. 7 shows such curves for 25, 20, 18 and 16% RDX. Comparison of calculated and measured failure diameters for these compositions are good in all cases except for 16% RDX (Table VII).

Table VII

Calculated and Measured D_f of RDX-Loaded PBAA Propellants

	D_f, in.	
% RDX	Measured	Calculated
25	1.00 - 1.15	1.12
20	1.61 - 1.75	1.81
18	2.25 - 2.50	2.52
16	2.75 - 3.00	4.62

Discussion

The large discrepancy in the calculated and measured failure diameter for the 16% RDX formulation indicates that Eq. 11 does not adequately describe this propellant system. Before any definitive statement can be made about the failure diameter of the base propellant the form of this equation must be determined through velocity measurements at several RDX concentrations.

A few general remarks can be made about the system, however. In the case of plastisol propellants the form of Eq. 8 implies the existence of a limiting failure diameter. Above this diameter (about 4 in.) Eqs. 6 and 8 do not intersect, so no solution is possible. In the case of PBAA-binder propellants, an equation of the form of Eq. 11 where $(U^o - U_f)/U^o$ increases, rather than decreases, with increasing D_f — that is, where the coefficient of D_f is negative instead of positive — a solution exists at all diameters.

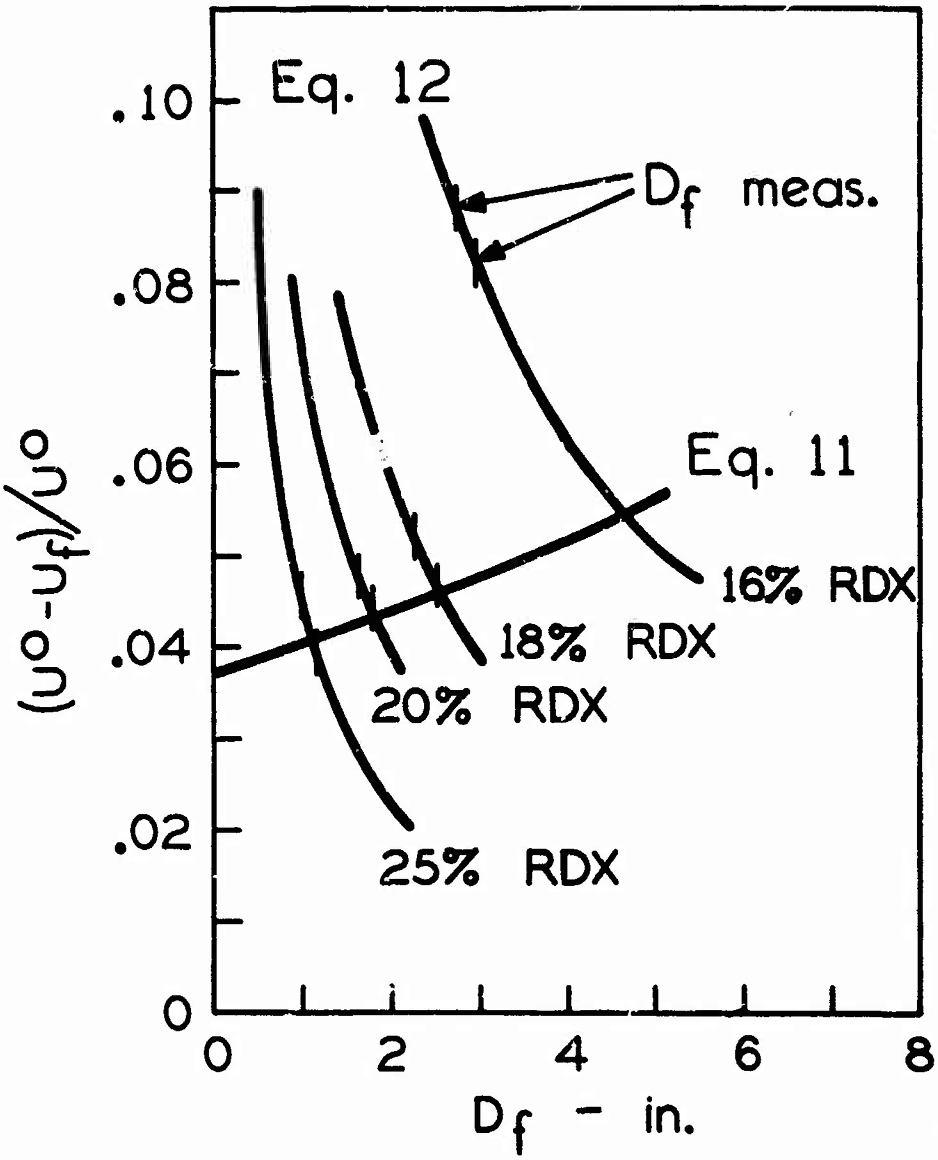

FIG. 7 CALCULATED RELATION BETWEEN D_f AND $(U^o - U_f)/U^o$ FOR PBAA COMPOSITE PROPELLANTS CONTAINING RDX.

With PBAA propellants another factor is limiting: the negative value of Q_2. If Eq. 6 is valid, Q must be positive. Therefore, when $n_1Q_1 + n_2Q_2 = 0$, detonation is no longer possible. This point corresponds to about 14% RDX when Eq. 11 is used to determine $(U^o - U_f)/U^o$. A similar result is obtained if $1/D_f$ is plotted as a function of RDX concentration. If a straight line is drawn through the data points, the failure diameter is infinite at ~10% RDX (Fig. 8).

The significance of the negative value for Q_2 can be seen by considering that this is an effective heat of reaction for this particular detonation reaction. If the ammonium perchlorate does not react in the detonation reaction zone, but instead reacts later, past the C-J plane, and

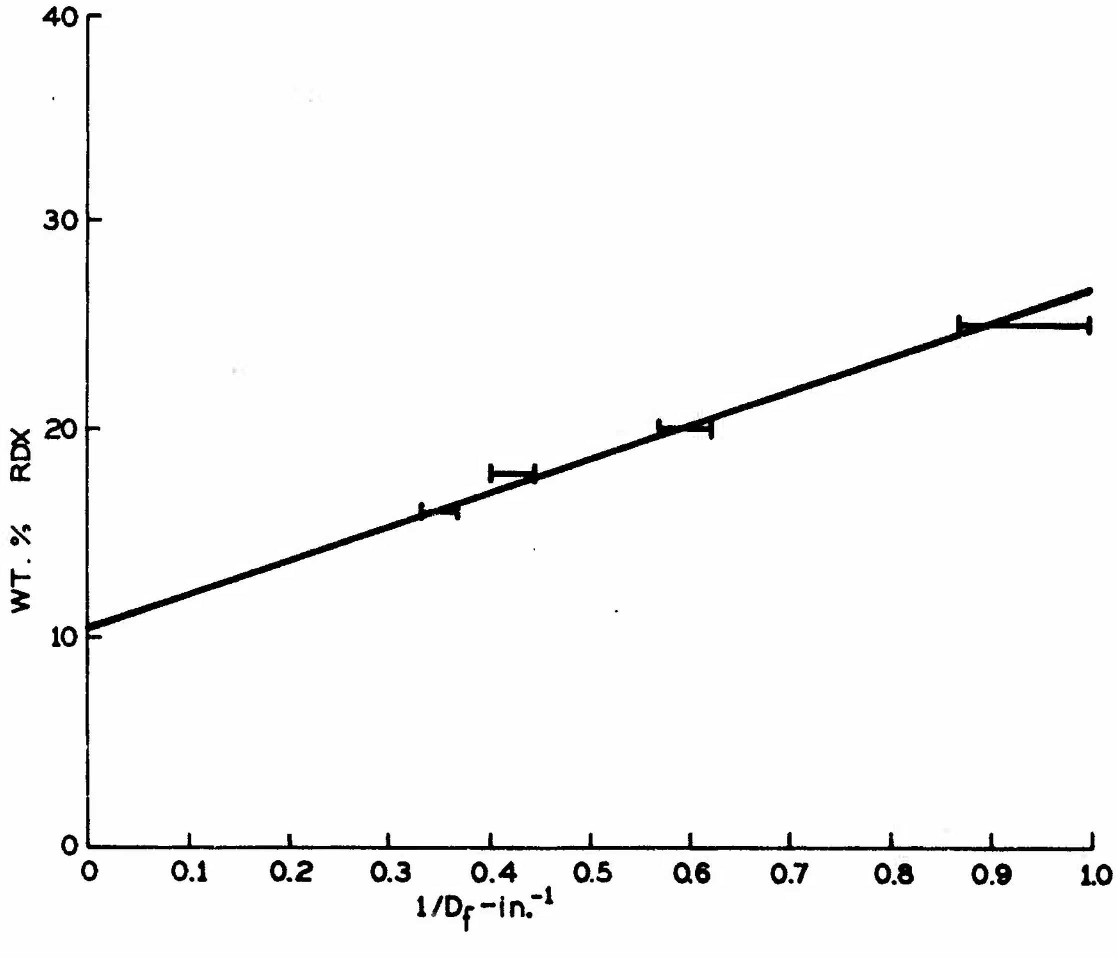

FIG. 8 INVERSE FAILURE DIAMETERS OF PBAA FORMULATIONS AS A FUNCTION OF RDX CONCENTRATION.

some of the energy released by the RDX is used to start reaction in ammonium perchlorate (or perform other work on the propellant), Q_2 would indeed appear as a negative quantity. This, in effect, would mean that the base propellant is incapable of detonation. The detonation reaction observed may be that of diluted RDX. This could be tested by using a base propellant with an inert salt substituted for the ammonium perchlorate.

CONCLUSIONS AND RECOMMENDATIONS

As originally proposed, this program had two phases. The first was to determine whether or not the application of the linear relation between failure diameter and change in detonation velocity (of the form of Eq. 8), derived from experiments with plastisol-nitrocellulose composite propellants, was valid for nonreactive-binder composite formulations. At this point, at least for the specific case of PBAA compositions, we must conclude that it is not. The second phase, predicated on such a negative conclusion, was to find some new relation between U^o, U_f, and D_f by conducting a series of tests on fuel-binder propellants at three or more RDX concentrations. It is clear now that Phase II should be carried out; however, it has not been, owing to exhaustion of the RDX supply in Phase I and the differences in results which can be expected with other lots of RDX in the light of now-known lot-to-lot variations (see Appendix B).

Although the results did not have the form of those we anticipated, the evidence still supports, however tenuously, the conclusion that the unadulterated fuel-binder propellant used here is not detonable. The apparent negative heat of detonation, Q_2, of the base propellant alone, together with the obvious requirement that the heat of detonation of the system, as a whole, Q, must be positive (Eqs. 6 and 7), implies that, as far as the detonation process is concerned, the base propellant is acted upon and makes no positive energy contribution. It is natural to ask whether RDX in a dummy propellant will have the same effects on failure diameter and detonation velocity as it does in "live" propellant. The absence of a significant difference in detonation characteristics between real and dummy propellants loaded with RDX would greatly strengthen the argument that the unadulterated forms are prima facie nondetonable.

Marked differences have been observed between failure diameters determined by the direct-match and the inverse techniques (Appendix A). Such differences may hold great significance as regards hazard evaluation of large solid propellant grains. Contrary to our experience with reactive-binder propellants, large charges of RDX-loaded PBAA composite propellants can be detonated by boosters of significantly smaller diameter than the direct-match failure diameter. Whether the magnitude of the difference in the failure diameters obtained by the two methods is constant or scales with charge size should be determined. Also, the extent of adulterant-concentration and particle-size effects should be explored.

The belated discovery of large differences in detonation characteristics among lots of RDX leaves us with an optimistic point of view. The levels of RDX concentration required to obtain small failure diameters in this investigation were much greater than those reported by Aerojet on Project SOPHY.[7] These high concentrations militate strongly against the assumption that the kinetic coefficient A in Eq. 6 is a function solely of the base propellant and invariant with RDX addition.. Information we have subsequently obtained about RDX (Appendix B) indicates the further work using currently produced RDX could be accomplished at much lower RDX levels. Although, in principle, the same result should be obtained in the limiting case of no RDX addition, the existence of lot-to-lot variation precludes the direct comparison of results obtained at different locations.

To recapitulate, we recommend

(1) estimation of the degree of participation of base propellant in the "true" detonation phenomenon by comparison of failure diameters of RDX-adulterated "live"- and dummy-oxidizer formulations, to learn whether the adulteration is a feasible approach to D_f prediction,

(2) continuation of efforts to find a valid relation between failure-diameter and diameter-dependence of detonation velocity of RDX-adulterated propellants, provided contribution by the base propellant is indicated in (1) above, and

(3) comparative parametric studies of direct-match and inverse methods of determining failure diameters, to make sure that latent fallacies do not exist in the conventional, booster-type of hazard evaluation.

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SYMBOLS LIST

t	Time
c_v	Specific heat at constant volume
R	Gas constant
T	Temperature
ν	Frequency factor
E_a	Activation energy
ξ_1	Detonation reaction zone length
D	Charge diameter
D_f	Failure diameter
U	Detonation velocity
U^o	Detonation velocity at infinite diameter
U_f	Detonation velocity at failure diameter
Q	Heat of detonation of entire system
Q_1	Heat of detonation of RDX
Q_2	Heat of detonation of base propellant
n_1	Weight fraction of RDX
n_2	Weight fraction of base propellant
A	$\dfrac{0.75 c_v R T^2}{\nu E_a} \exp\left(\dfrac{E_a}{RT}\right)$

APPENDIX A

Formulation Studies

The first step in this investigation was to select two propellant formulations which had reasonable failure diameters, less than 2.5 inches, and differed in RDX concentration by at least 5 numerical per cent. A further restriction was that the formulations have a constant base with added RDX, thereby giving a variable solids loading; since RDX is stoichiometrically balanced itself, this method allowed preservation of the over-all propellant stoichiometry regardless of RDX level.

The first formulation examined was RH-B-14,a plasticized PBAA formulation with 10% RDX (Table A-I). This propellant was capable of being manufactured in a turbine mixer. It was found that the failure diameter was too large, >3.00 in. (Table A-II), and that the resulting formulation did not approximate a usable propellant because it was too soft.

Another series of formulations was then examined, RH-B-15 through RH-B-19 (Table A-I), with larger concentrations of RDX. The base formulation was tailored so that the mix viscosity of 30% RDX formulation, RH-B-19 would make it castable. The failure diameters of these formulations were determined by the so-called inverse technique* (Table A-II). This technique employs an oversize acceptor and the diameter of the donor is varied to determine, within limits, what diameter donor charge is required to give a 50% probability of initiating a detonation in the acceptor. The donor diameter so determined is taken as an approximation to the failure diameter of the acceptor. With this failure-diameter criterion the formulations RH-B-19, 30% RDX, and RH-B-18, 20% RDX, were chosen as acceptable formulations. However, when these formulations were cast into rounds to

*W. W. Brandon, "Importance of Flexibility in Gap Sensitivity Testing", Bull. 16th Mtg. JANAF Solid Prop. Group V, 109 (1960).

Table A-I

Propellant Formulations

Non-Plasticized Formulations, RH-B-

Ingredient, Wt. %	15[a]	16	17	18	19	22	23	24	25
APC[b]	62.5	53.2	46.9	50.0	43.8	45.6	52.5	56.3	51.25
AL[c]	12.5	10.6	9.4	10.0	8.7	9.1	10.5	11.2	10.25
PBAA/ERL 2774[d]	25.0	21.2	1[illegible].7	20.0	17.5	18.3	21.0	22.5	20.50
RDX	0	15.0	25.0	20.0	30.0	27.0	16.0	10.0	18.00

Plasticized Formulations, RH-B-

Ingredient, Wt. %	14	20	21
APC[b]	52.94	40.0	45.72
Al[c]	10.59	8.5	9.71
PBAA/ERL 2774[d]	21.18	18.5	21.14
DOP[e]	5.29	----	----
DOA[f]	----	3.0	3.43
RDX	10.00	30.0	20.0

Carboxy-Terminated Polybutadiene Formulation

Ingredient, Wt. %	RH-C-33
HC[g]	18.5
APC[b]	40.0
Al[c]	8.5
DOA	3.0
RDX	30.0

[a] base composition
[b] cc/ce = 70/30 ammonium perchlorate (see Fig. A-3)
[c] 3-5μ aluminum [Alcoa 140]
[d] PBAA/ERL 2774 = 89.5/10.5 [ERL 2774 = epoxy resin (Union Carbide)]
[e] dioctyl phthalate
[f] dioctyl adipate
[g] ZL 434/ERLA/MAPO = 95.16/1.28/3.56 [ZL 434 = carboxy-terminated polybutadiene polymer (Thiokol Chemical Corp.); ERLA = trifunctional epoxy resin (Union Carbide); MAPO = tris[1-(2-methyl)aziridinyl]-phosphine oxide (Interchemical Corp.)].

Table A-II

Consolidated Failure - Diameter Data for Class A-RDX-Containing Fuel-Binder Composite Propellants

Designation	Batch	% RDX	Total No. of Shots	Acceptor Diam., in.	Donor Diam., in.	D_f, in.[a]
RH-B-15[b]	1000	0	2	3	3	>3.00
RH-B-14[c]	1000	10	2	3	3	>3.00
RH-B-16	1000	15	2	3	3	>3.00
RH-B-18	1000	20	4	3	d	1.61 - 1.75
RH-B-18	1002		4	3	d	1.75 - 1.88
RH-B-18	1002		2	e	e	2.00 - 2.50
RH-B-21[f]	1000		2	3	3	>3.00
RH-B-17	1000	25	2	3	d	1.00 - 2.07
RH-B-22	1000	27	4	1.38	2	>1.38
RH-B-19	1000	30	6	2	d	1.00 - 1.15
RH-C-33	1000		1	1.25	1.25	>1.25
RH-B-20	1002		1	1.25	1.25	>1.25
RH-B-20	1003		3	e	e	1.25 - 1.50

[a] direct-match method, unless otherwise indicated
[b] uncured propellant slurry
[c] turbine-mixed DOP-plasticized composition
[d] inverse technique
[e] matched donor-acceptor
[f] DOA-plasticized composition

check the failure diameters two difficulties became apparent: (1) Contrary to wide experience with reactive-binder propellants the inverse technique gave D_f values which were much smaller than those found by the normal direct-match method whereby the acceptor diameter (≤ donor diameter) is reduced until go—no go limits are found. (2) The mix viscosity of the RH-B-19 was so high that void-free charges less than 1 in. in diameter were impossible to cast with the casting techniques then employed.

During this period of the investigation several parameters were investigated which included formulational variations and different casting procedures.

DOA (dioctyl adipate) was added to the base formulation to solve the casting problems in the small-diameter charges. Therefore, charges were cast of RH-B-20 (30% RDX) and RH-B-21 (20% RDX). But the addition of plasticizer did not solve the casting problems. The rounds were cast in a vacuum chamber and the problem of voids was caused by the propellant degassing during the casting, even though the propellant had been vacuum mixed for a minimum of 30 minutes. Holding the vacuum chamber at about 100 mm Hg permitted the propellant to flow through the slits into the cardboard casting tubes with no gassing. However, the slit-deaeration step was omitted with diameters less than one inch because otherwise the flow of the propellant was too slow to make the casting of these smaller charges feasible. This limited the choice of formulations to those having failure diameters of greater than one inch.

A carboxyterminated polybutadiene formulation RH-C-33, 30% RDX, was cast to determine if this propellant type had any advantage over the polybutadiene—acrylic acid formulations. Since the former had a longer cure time and offered no apparent improvement, further formulational studies with the RH-C propellants were abandoned.

Owing to its availability, Class A RDX had been used in the initial formulation studies. Subsequently, substitution of the much smaller particle size Class E RDX markedly decreased the failure diameter at a given RDX concentration (Tables A-II and A-III). The lots used in these experiments had weight-median-diameter particle sizes of 200μ (Class A) [wet screen] and 16μ (Class E) [Micromerograph]; the complete particle-size distributions are shown in Figs. A-1 and A-2. (The particle size distribution of the ammonium perchlorate used is shown in Fig. A-3.)

Table A-III

Consolidated Failure-Diameter Data for Class E-RDX-Containing PBAA Propellants

Designation	Batch No.	% RDX	D_f, in.[a]	RDX Lot
RH-B-24	1000	10	> 2.5	E22-A
RH-B-23	1000	16	> 1.25[b]	E22-A
	1001		2.75 - 3.0	E22-A
RH-B-25	1000[c]	18	2.25 - 2.50	E22-B
	1001[c]		2.25 - 2.50	E22-B
	1002[c]		2.25 - 2.50	E22-B
	1003[c]		2.25 - 2.50	E22-B
	1004[c]		< 2.0	d
	1005[c]		< 2.0	E20-B
	1006[c]		> 2.5	E22-C
	1007[c]		2.25 - 2.50	E22-C
	1008		0.95 - 1.05[e]	E20-B
RH-B-18	1003	20	< 1.50	E20-B
	1003		< 1.00[f]	E20-B
	1004		1.61 - 1.75	E22-A
RH-B-17	1001	25	1.00 - 1.15	E22-A
	1002[c]		1.00 - 1.15	E22-A
	1003[c]		1.00 - 1.15	E22-B

[a] direct-match method, unless otherwise indicated
[b] 2"-diam. booster
[c] batches which include velocity rounds
[d] 42% E22-B, 58% E20-B
[e] see footnote , p. A-9
[f] inverse technique on 3" acceptors

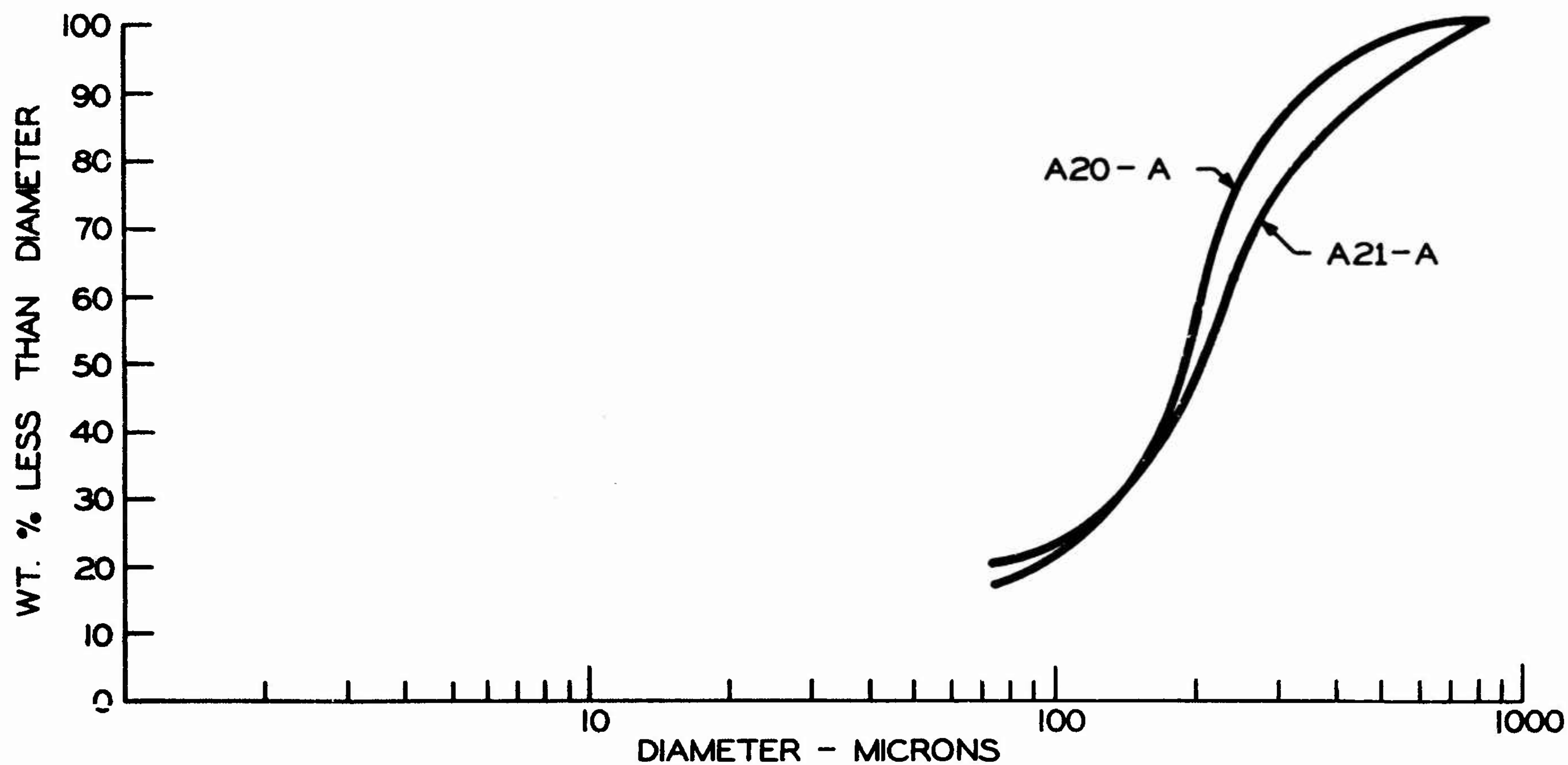

FIG. A-1 PARTICLE-SIZE DISTRIBUTION OF TYPE B CLASS A RDX (wet-screen analysis).

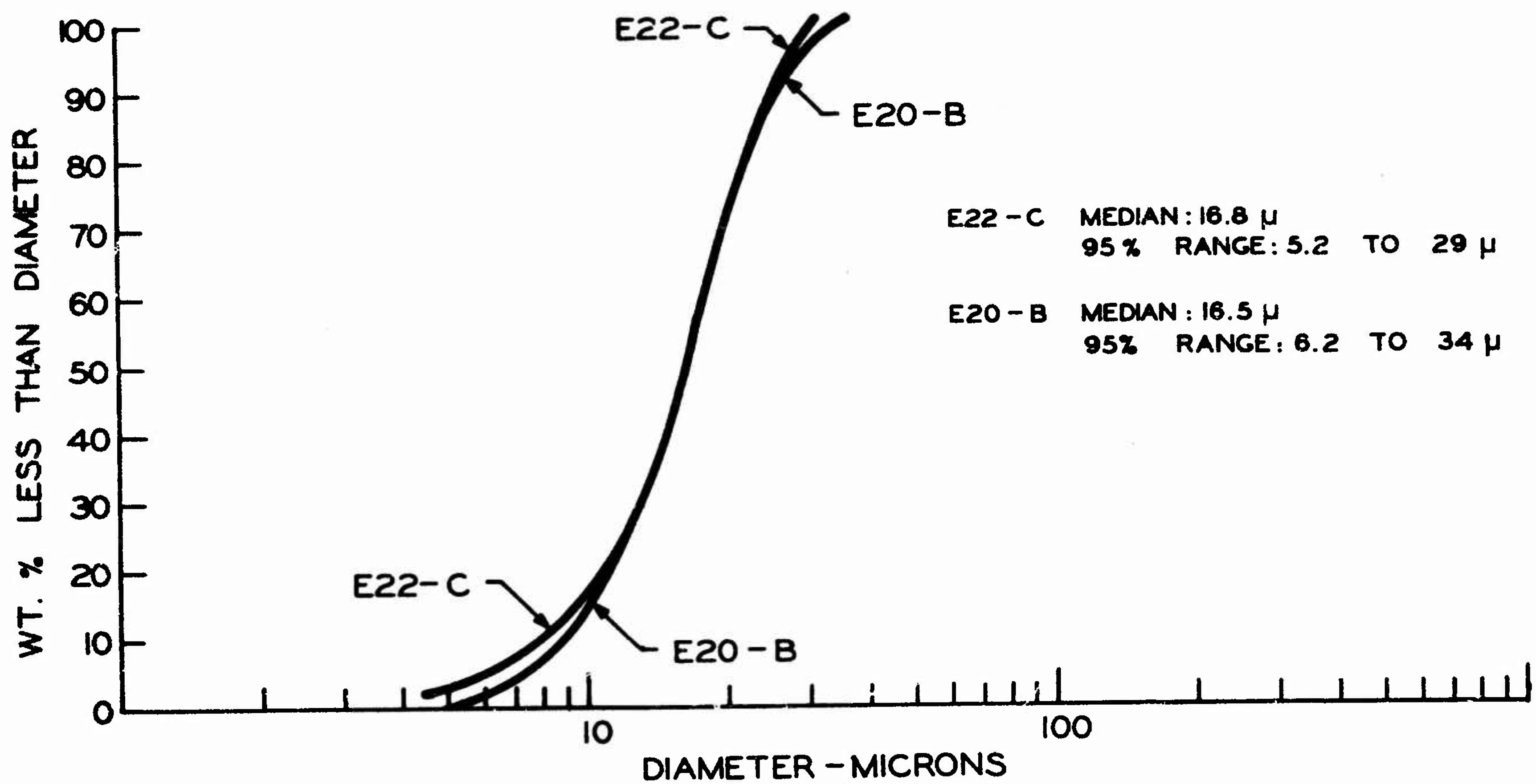

FIG. A-2 PARTICLE-SIZE DISTRIBUTION OF TYPE B CLASS E RDX (micromerograph).

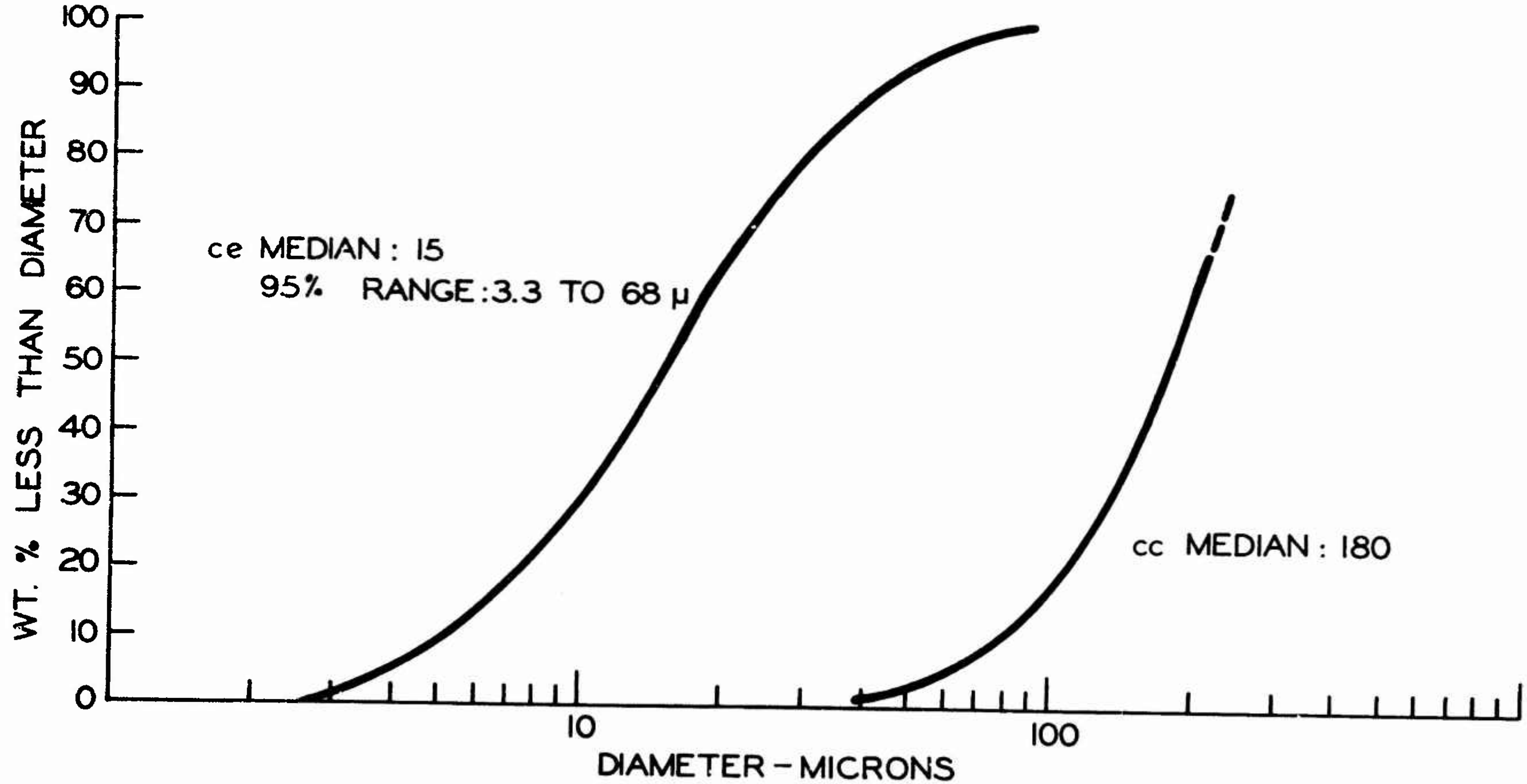

FIG. A-3 PARTICLE-SIZE DISTRIBUTION OF AMMONIUM PERCHLORATE (micromerograph).

The failure diameters of four formulations containing Class E RDX were determined by the direct-match method. The occurrence of a detonation was determined by perforation of a mild steel witness plate located at the distal end of the acceptor. This criterion however was sometimes ambiguous; in such cases another trial was made, with a duplicate charge, and the event was photographed with a streak camera (Beckman & Whitley Model 194). A detonation was indicated by a luminous front of constant velocity for the entire length of the acceptor. From these failure-diameter data (Table A-III) the formulations RH-B-17 (25% Class E RDX and and RH-B-25 (18% Class E RDX) were chosen for velocity determinations.

During the course of the casting of the velocity charges four lots of Class E RDX were used, Rohm & Haas designations E22-A, E22-B, E22-C and E20-B. The last letter was assigned, in alphabetical order, for each lot portion of RDX dried. The drying process apparently did not affect the failure diameter or the detonation velocity of the resulting propellant (compare Batches 1002 and 1003 of RH-B-17). (Here "lot" refers to raw material while "batch" refers to finished propellant.) However, it was found that E20-B gave a lower failure diameter than E22-A when compounded into RH-B-18 (compare Batches 1003 and 1004). The existence of the effects of lot-to-lot variation was not known at the time that velocity rounds of RH-B-25 were cast. As a result E20-B was used while another portion of E22-B was being dried. The subsequent discovery of the difference in D_f negated results obtained from Batches 1004 and 1005. The magnitude of the shift in D_f was determined later from a supplementary batch of RH-B-25 made with E20-B (Batch 1008). From a D_f between 2.25 and 2.50 in. with E22 the value decreased to between 0.95 and 1.05 in. with E20*.

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*When a "go" was obtained with a $1\frac{1}{4}$-in.-diam. round (the smallest castable diameter), oversize rounds were sliced into $\frac{1}{2}$-in.-thick wafers, reduced in diameter by a cork-borer-type cutter, and the wafers were then stacked poker-chip fashion. With charges formed in this manner the failure diameter of RH-B-25-1008 was $0.95" < D_f < 1.05"$. The validity of this method was checked by sectioning surplus charges of RH-B-20-1003 (30% Class A RDX), 1.25 in. and 1.50 in. in diameter (the established go—no-go limits for consolidated rounds), and restacking the resulting wafers without a reduction in diameter. These charges, when fired, indicated no change in D_f. Similarly, RH-B-23-

RDX lots E20-B and E22-C were analyzed for HMX content. Lot E20-B, which gave the lower failure diameter, had over four times as much HMX (8.4%) as Lot E22-C (2.0%). In view of the close similarity in chemical and explosive characteristics of HMX and RDX, the mere presence of the different amounts of HMX is, in itself, not significant.* The real importance of the concentration difference is the indication of a significant difference in the manufacture and processing of the RDX lots. Unpublished work by Lawrence Radiation Laboratory has shown that failure diameters of pure solid explosives are extremely sensitive to changes not only in the size and shape of the particle aggregate but also in its crystalline substructure. Accordingly the two lots of RDX were then examined microscopically. As Fig. A-4 shows, the crystals were different in shape as well as size. The photomicrographs indicate that the Micromerograph analyses are in error. Since the Micromerograph technique is predicated on the particles' being of nearly spherical shape, the rod-like form of E22 would give spurious low values. The shift in propellant failure diameter is easily accounted for, at least qualitatively, by the difference in true RDX particle size for the propellants containing the two lots. However, the possibility cannot be ruled out that some other property of the crystals is contributing to the change in failure diameter.

Distance—time measurements for all velocity rounds are assembled in Table A-IV. However, for the several reasons mentioned above, the results for rounds from RH-B-19-1001, RH-B-25-1004 and 1005 have not been considered in treating the data.

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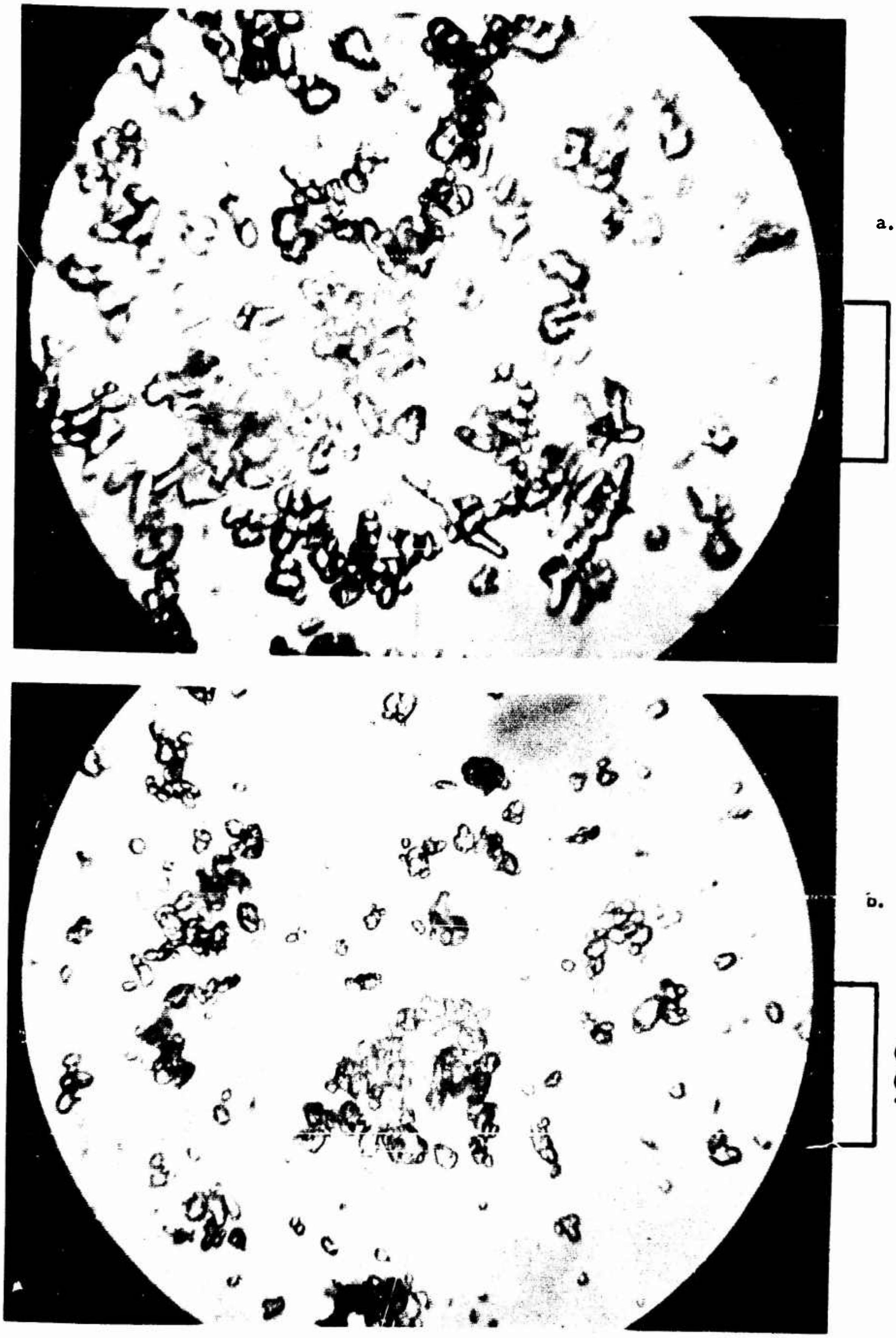
*Mr. Edward James, Jr., Lawrence Radiation Laboratory, private communication, May 1965.

1001 (16% Class E RDX) and RH-B-25-1007 (18% Class E RDX) showed no difference in D_f observed with integral and reconstituted rounds. The validity of the method was further confirmed with a plastisol-nitrocellulose composition in which charges were formed of wafers cut not only from cylindrical rounds but also from a cast 1/4-in.-thick sheet.



a. E22

b. E20

FIG. A-4 PHOTOMICROGRAPHS OF TYPE B CLASS E RDX LOTS.

Table A-IV

Consolidated Velocity Data

Round Designation	Diam., (in.)	Length, (in.)	Probe Location, in. from Donor	Time,[a] (μsec)	Δt,[a] (μsec)	Time,[b] (μsec)
RH-B-19-1001	2.5	$34\frac{3}{8}$	2	Trigger[c]		Trigger
			8	$26\frac{4}{8}$		
			14	$53\frac{2}{8}$	$26\frac{6}{8}$	
			20	$79\frac{2}{8}$	26	
			26	$105\frac{6}{8}$	$26\frac{4}{8}$	
			32	$132\frac{1}{8}$	$26\frac{3}{8}$	131
RH-B-19-1001	2	$33\frac{1}{2}$	1.5	Trigger[c]		Trigger
			7.5			
			13.5	Failed to Trigger		
			19.5			
			25.5			
			31.5			134.7
RH-B-17-1002-7	1.25	$34\frac{1}{8}$	3	$13\frac{5}{8}$		
			10	$45\frac{7}{8}$	$32\frac{2}{8}$	
			17	$78\frac{1}{8}$	$32\frac{2}{8}$	
			24	$110\frac{3}{8}$	$32\frac{2}{8}$	
			31	$142\frac{4}{8}$	$32\frac{1}{8}$	
RH-B-17-1002-8	2.0	34	3	13		
			10	45	32	
			17	$76\frac{7}{8}$	$31\frac{7}{8}$	
			24	$108\frac{5}{8}$	$31\frac{6}{8}$	
			31	$140\frac{1}{8}$	$31\frac{4}{8}$	

Table A-IV continued

Round Designation	Diam., (in.)	Length, (in.)	Probe Location, in. from Donor	Time,[a] (µsec)	Δt,[a] (µsec)	Time,[b] (µsec)
RH-B-17-1002-9	2.50	34	3	13 4/8		
			10	45 6/8	32 2/8	
			17	76 7/8	31 1/8	
			24	108 5/8	31 6/8	
			31	139 6/8	31 1/8	
RH-B-17-1002-10	4.0	31 3/8	4	18 3/8		
			10		53 7/8	
			16	72 2/8		
			22	99	26 6/8	
			28	126	27	
RH-B-17-1003-7	1.25	33 1/2	3	14		
			10		63 5/8	
			17	77 5/8		
			24	110	32 3/8	
			31	142 2/8	32 2/8	
RH-B-17-1003-8	2.00	34	3	13 6/8		
			10	45 4/8	31 6/8	
			17	76 7/8	31 3/8	
			24	108 5/8	31 6/8	
			31	139 6/8	31 1/8	
RH-B-17-1003-9	2.5	32 5/8	3	13 2/8		
			10	45 2/8	32	
			17	76 5/8	31 3/8	
			24	107 7/8	31 2/8	
			31	139 6/8	31 7/8	

Table A-IV continued

Round Designation	Diam., (in.)	Length, (in.)	Probe Location, in. from Donor	Time,[a] (μsec)	Δt,[a] (μsec)	Time,[b] (μsec)
RH-B-25-1000-11	4	39	3	$14\frac{2}{8}$		
			11.5	$55\frac{5}{8}$	$41\frac{3}{8}$	14.6
			20	$97\frac{1}{8}$	$41\frac{4}{8}$	
			28.5	$138\frac{4}{8}$	$41\frac{3}{8}$	
			37	$179\frac{5}{8}$	$41\frac{1}{8}$	
RH-B-25-1001-11	2.5	$40\frac{3}{4}$	3	$13\frac{7}{8}$		14.0
			12	58	$44\frac{1}{8}$	
			21	$103\frac{1}{8}$	$45\frac{1}{8}$	
			30	148	$44\frac{7}{8}$	
			39	$192\frac{5}{8}$	$44\frac{5}{8}$	191.9
RH-B-25-1001-12	3.0	$40\frac{3}{8}$	3	$13\frac{2}{8}$		14.3
			12	$57\frac{6}{8}$	44	
			21	102	$44\frac{2}{8}$	
			30	$146\frac{1}{8}$	$44\frac{1}{8}$	
			39	191	$44\frac{7}{8}$	191.4
RH-B-25-1002-11	6.0	$36\frac{3}{4}$	3	14		15.7
RH-B-25-1003-11			11	$50\frac{4}{8}$	$36\frac{4}{8}$	
			19	$91\frac{4}{8}$	41	
			27	$129\frac{5}{8}$	$38\frac{1}{8}$	
			35	$168\frac{6}{8}$	$39\frac{1}{8}$	169.2
RH-B-25-1004-8	2.5	$41\frac{1}{4}$	3	14		13.5
			12	$58\frac{2}{8}$	$44\frac{2}{8}$	
			21	$102\frac{1}{8}$	$43\frac{7}{8}$	
			30	147	$44\frac{7}{8}$	
			39	191	44	190.9

Table A-IV continued

Round Designation	Diam., (in.)	Length, (in.)	Probe Location, in. from Donor	Time,[a] (μsec)	Δt,[a] (μsec)	Time,[b] (μsec)
RH-B-25-1004-9	3.0	40 7/8	3	13 2/8		14.2
			12	57 4/8	44 2/8	
			21	101 5/8	44 1/8	
			30	145 5/8	44	
			39	189	43 3/8	189.3
RH-B-25-1005-8	4.0	40 1/2	No Trigger			
RH-B-25-1006-9	2.5	40 5/8	No Detonation			
RH-B-25-1006-10	3.0	40 1/2	3	13 7/8		13.7
			12	58 1/8	44 2/8	
			21	103	44 7/8	
			30	147 2/8	44 2/8	
			39	191 7/8	44 5/8	191.4
RH-B-25-1007-9	4	41	3	13		
			12	57 7/8	44 7/8	
			21	101 4/8	43 5/8	
			30	146 5/8	45 1/8	
			39	189 2/8	42 5/8	

[a]Model 471 8mc Counter Chronograph, Potter Instrument Co., Inc., Great Neck, New York.
[b]Model 6146 EPUT® and TIMER, Beckman Instruments, Inc., Berkley Division, Richmond, California.
[c]Unless otherwise specified, trigger is at booster-charge interface.

APPENDIX B

Manufacture of RDX

In connection with our concern over the shift in failure diameters of RDX-containing propellant when compounded with different lots of Type B, Class E RDX a visit was made to Holston Army Ammunition Plant, Kingsport, Tennessee. Our analysis of RDX for HMX for the two RDX lots in question gave values of 8.4% and 2.0% HMX. This came as a surprise to the people at Holston. Their two-sigma limit for HMX in RDX is 5-14%. However, since our lot of RDX which showed 2% HMX could have been as much as 10 years old the following hypothesis was formulated. In the past 10 years several process changes have been made and it was not the practice 10 years ago to analyze RDX for HMX. Therefore the 2% analysis is reasonable and indicates a change in process history which could in turn affect failure diameter of RDX-containing propellants.

Some representative particle-size data on recent batches of Class E RDX are shown in Table B-I. The 5%-95% range for different lots has been found to be 5-54μ. All these analyses meet the specification of Class E RDX that 97% shall pass through a U. S. Standard Sieve Number 325.*

Table B-I

Comparison of Particle Size Determination of Class E RDX (Holston Data)

Method	Range of Median for Different Lots
Buckbee-Mears	7-31μ
Photoextinction	25-31μ
Sub Sieve Sizer	10.2-13.0μ

*"RDX, HMX, and Explosive Compositions", Technical Data Bulletin TD-2 Holston Army Ammunition Plant, Kingsport, Tennessee, April 1965.

During a plant tour the following process details were brought out:

(1) Classes A, B, C, and D RDX are recrystallized from cyclohexanone, Classes E and F from acetone.

(2) The recrystallization is carried out in 2000 lb. lots in which the solvent—RDX solution is added to a solvent—water solution under controlled temperatures. The particle size, and therefore the class, is determined by the recrystallization temperature and, since the temperature depends somewhat on the weather, the size distribution may be affected.

(3) The solvent is stripped off the RDX—solvent—water mixture, leaving a water slurry of RDX which is decanted and filtered into stainless steel bins. Some classification of particle size should be expected during this step of manufacture.

(4) The RDX is then shoveled, wet, from the stainless steel bins into cloth bags and placed in drums. Here further classification is not unlikely, since the first bag contains RDX mostly from the top of the bin. However, since the slurry does not flow, subsequent shovelfuls are taken cross-sectionally from top to bottom of the bin minimizing the classification in bags filled later from that bin.

(5) Ten pounds of isopropanol is then added to each drum as an antifreeze. The drum is then sealed and shipped.

(6) A lot of RDX is nominally 2000 lbs.; however, three lots can be cross-blended at Holston Army Ammunition Plant in order to give one lot of approximately 6000 lbs.

(7) The RDX which does not meet sieve size specifications (the only criterion other than purity) is used in the manufacture of Composition B. This includes "heels" which are high in HMX content. Composition B is therefore variable in HMX content and can be as high as 40% HMX.

In conclusion one can expect lot-to-lot variations in particle sizes of the RDX as manufactured by Holston Army Ammunition Plant which may be significant for experimentation involving failure diameters of RDX-containing propellant. One may also expect a slight particle size classification from bag to bag of the RDX as shipped. Furthermore, Composition B, as manufactured by Holston Army Ammunition Plant, should not be used as a standard explosive since its composition is variable in regard to particle size as well as HMX content.